Possible role of ^3He impurities in solid ^4He

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Abstract. - We use a quantum lattice gas model to describe essential aspects of the motion of ^4He atoms and of ^3He impurities in solid ^4He. This study suggests that ^3He impurities bind to defects and promote ^4He atoms to interstitial sites which can turn the bosonic quantum disordered crystal into a metastable supersolid. It is suggested that defects and interstitial atoms are produced during the solid ^4He nucleation process where the role of ^3He impurities (in addition to the cooling rate) is known to be important even at very small (1 ppm) impurity concentration. It is also proposed that such defects can form a glass phase during the ^4He solid growth by rapid cooling.

Kim and Chan [1] (KC) using the torsional oscillator technique found a decrease in the resonant period of solid ^4He confined in porous vycor glass and in bulk solid helium below $200mK$, indicating the possible onset of superfluidity in solid helium. The experimental results of KC have been independently confirmed by Rittner and Reppy [2] (RR) using a different geometry to confine the solid. In addition, RR observed that the superfluidity of solid helium can be significantly influenced or eliminated by annealing of the solid helium sample. Because of these history-dependent results and the negative results in attempts to drive flow by pressure [3], the interpretation of the results of KC is subject to debate.

Furthermore, it has been observed that the superfluid response and superfluid fraction are strongly dependent on the amount of isotopic ^3He impurities which exist in the naturally available helium. When, for example, the naturally occurring concentration of ^3He impurities of about one part per million is reduced to less than one part per billion [4] the superfluid fraction is reduced from about 1% to approximately 0.03%. When the ^3He impurity concentration is increased to about 0.1% the superfluid fraction vanishes [1]. Previously, Ho *et al.* [5] found an anomalous behavior of the acoustic attenuation and velocity in solid ^4He below $\sim 200mK$ at a low concentration of ^3He impurities.

The possibility of superfluidity of solid ^4He has been extensively discussed [6–9]. It is of fundamental value for our understanding of this and related phenomena because of the implication that there can be coexistence between spatial and momentum space order [10].

In this paper we study the role of very low density of impurities in a bosonic hard core solid. While the theoretical studies of the role of impurities in solid helium has a long history [6, 11], the present study, based on an analogy with models of doped quantum antiferromagnets and by using rather recently developed techniques for such systems, sheds some "new" light on the problem. Our model, that describes the impurity motion in an otherwise ideal quantum bosonic crystal, maps to a quantum spin model with antiferromagnetic (AF) coupling and AF order in one direction and ferromagnetic coupling in the perpendicular direction with impurities moving through the lattice. The impurity motion between sub-lattices couples to quantum fluctuations of these pseudo-spin degrees of freedom which correspond to the boson hopping from an occupied site of the solid to an empty interstitial site. We find that, for the limit which describes the case of solid ^4He, interstitial atoms are well-defined delocalized excitations. It is suggested that during the ^4He solid nucleation process, the ^3He impurities stabilize point defects such as dislocations or disclinations at the solid-to-liquid interface which become lines of defects as the solid grows. These defects can become highly entangled and can form a glass phase at low temperature. The ^3He impurities become bound to these defects and promote mobile interstitial ^4He atoms into the disordered solid. We find that such interstitial atoms can condense and may be responsible for the ob-

served behavior [1].

Let us consider a lattice gas model to describe the bosonic solid and the added impurities. In such a model, we need to consider the interstitial sites as part of the lattice and, thus, the ideal quantum solid containing no vacancies and no impurities, corresponds to a fractionally occupied lattice. For example, the ideal triangular solid corresponds to the case of 1/3 filling, namely, to a $\sqrt{3}\times\sqrt{3}$ ordered solid and the ideal square lattice solid corresponds to the $\sqrt{2}\times\sqrt{2}$ checkerboard solid, i.e., 1/2 filling of the lattice with bosons. Our model Hamiltonian describing a bosonic quantum solid (such as solid ^4He) and a small concentration of impurities (such as ^3He atoms) may be written as follows:

$$\hat{\mathcal{H}} = \hat{\mathcal{H}}_B + \hat{\mathcal{H}}^* + \hat{\mathcal{H}}_{int} - \mu\sum_i \hat{N}_i - \mu^*\sum_i \hat{n}_i, \quad (1)$$

$$\hat{\mathcal{H}}_B = \sum_{<ij>}[-t_b(B_i^\dagger B_j + h.c.) + V\hat{N}_i\hat{N}_j] + U\sum_i \hat{N}_i^2, \quad (2)$$

$$\hat{\mathcal{H}}^* = \sum_{<ij>}[-t(c_i^\dagger c_j + h.c.) + W\hat{n}_i\hat{n}_j] + U^*\sum_i \hat{n}_i^2, \quad (3)$$

$$\hat{\mathcal{H}}_{int} = V^*\sum_{<ij>}(\hat{N}_i\hat{n}_j + \hat{n}_i\hat{N}_j) + U'\sum_i \hat{n}_i\hat{N}_i, \quad (4)$$

$\mathcal{H}_B$ is the Hamiltonian of the bosonic solid, and $B_i^\dagger(B_i)$ are boson creation (annihilation) operators and $\hat{N}_i = B_i^\dagger B_i$. The fermion operators $c_i^\dagger(c_i)$ create (or annihilate) impurities on the site i and we have suppressed their spin degree of freedom for simplicity. Here, n_i is the impurity number operator. We will consider the $U\to\infty$, $U^*\to\infty$ and $U'\to\infty$ limits (single-site occupation subspace) because of the the hard-core interaction. V, V^* and W are positive because of the additional energy cost to place an atom in an interstitial site.

In the absence of impurities the well-known pure bosonic Hamiltonian (2), in the limit of $U\to\infty$ and under the transformation $B_i^\dagger \to S_i^+, B_i \to S_i^-, N_i \to S_i^z - \frac{1}{2}$, where $(\hat{S}_i^x, S_i^y, S_i^z)$ are spin-1/2 operators, reduces to the anisotropic spin-1/2 Heisenberg model [12]

$$\hat{\mathcal{H}}_B = \sum_{<ij>}[JS_i^zS_j^z + J_{xy}(S_i^xS_j^x + S_i^yS_j^y)] - H\sum_i S_i^z, \quad (5)$$

where $J = V > 0$ and $J_{xy} = -2t_b < 0$ and $H = \mu - z/2V$, z is the coordination number.

In order to illustrate the effects of the impurities on the stability of the quantum solid and, vice versa, we first consider the square lattice because of its simplicity. Following the general spin-wave (SW) theory for an ordered square lattice quantum antiferromagnet [12], we separate the ordered square lattice in two sub-lattices, A (or up, or occupied) and B (down, or empty) and we consider boson operators $a_i^\dagger$ and $b_i^\dagger$ which create spin-deviations with respect to the classical Néel ground state in sites of the corresponding sub-lattice. The Hamiltonian (5) is approximated by keeping terms up to quadratic in spin-deviation operators; using the Fourier transforms $a_{\mathbf{k}}$ and $b_{\mathbf{k}}$ of the operators a_i and b_i (as defined in Ref. [12]), where $\mathbf{k}$ takes values from the Brillouin zone of the $\sqrt{2}\times\sqrt{2}$ sub-lattice and introducing the Bogoliubov canonical transformation, $a_{\mathbf{k}} = u_k\alpha_{\mathbf{k}} + v_k\beta_{-\mathbf{k}}^\dagger$, $b_{\mathbf{k}} = u_k\beta_{\mathbf{k}} - v_k\alpha_{-\mathbf{k}}^\dagger$, where $\alpha_{\mathbf{k}}^\dagger$ and $\beta_{\mathbf{k}}^\dagger$ are boson creation operators, the Hamiltonian takes the form

$$\mathcal{H}_L^B = E_0 + \sum_{\mathbf{k}}\Big(\omega_{\mathbf{k}}^\alpha\alpha_{\mathbf{k}}^\dagger a_{\mathbf{k}} + \omega_{\mathbf{k}}^\beta\beta_{\mathbf{k}}^\dagger\beta_{\mathbf{k}}\Big), \quad (6)$$

where $\omega_{\mathbf{k}}^{\alpha,\beta} = dJ\epsilon_{\mathbf{k}} \pm H$, $\epsilon_{\mathbf{k}} = \sqrt{1-\lambda^2\gamma_{\mathbf{k}}^2}$, $\gamma_{\mathbf{k}} = 1/2(cos(k_x) + cos(k_y))$, $d = 2$ and $\lambda = J_{xy}/J$, and $u_{\mathbf{k}} = \sqrt{1/2(1/\epsilon_{\mathbf{k}}+1)}$, $v_{\mathbf{k}} = -sgn(\gamma_{\mathbf{k}})\sqrt{1/2(1/\epsilon_{\mathbf{k}}-1)}$.

It follows from Eq. 6 that for $\omega_0^{\alpha,\beta} \geq 0$, i.e., for $H \geq dJ\sqrt{1-\lambda^2}$, the Néel ordered ground state is unstable. Ferromagnetic (superfluid in the Bose system) order develops in the xy direction and the spins are canted in order to acquire a component along the direction of the field. The phase diagram obtained for this model with this spin-wave approximation agrees reasonably well with that obtained by other techniques. [13] When the square lattice is half-filled there is a gap $G = dJ\sqrt{1-\lambda^2}$ for creating a propagating pseudo-spin wave excitation, i.e., to promote a boson atom to the interstitial band. These interstitial quasiparticles move in a band which in the limit

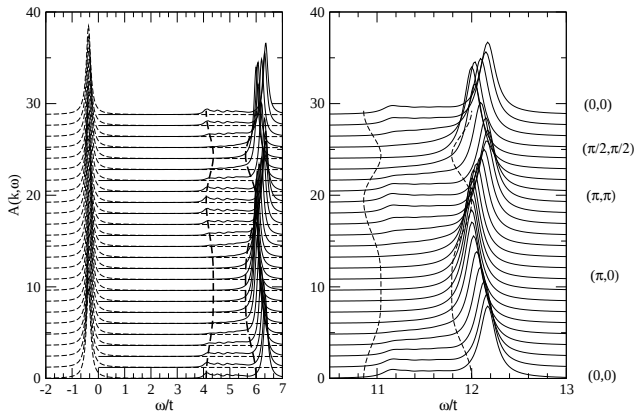

Fig. 1: The spectral functions $A(\mathbf{k},\omega)$ for $J/t = 3$ (left) and $J/t = 6$ (right) along the Brillouin zone path $(0.0) \to (\pi,0) \to (\pi,\pi) \to (0,0)$.

We wish to extend this approach to study the motion of a single impurity inside the quantum solid. As in Ref. [14], we consider as reference state the Néel state with an impurity on the A (substitutional) or B (interstitial) sublattices. We imagine that there exist operators $h_i^\dagger$ and $f_i^\dagger$ which operate on the Néel state and replace respectively an up-spin or down-spin with an impurity. For the single impurity case and by keeping only up to linear terms in spin-deviation operators the terms given by Eqs. (3) (4) in terms of these operators take the form $\hat{\mathcal{H}}^* + \hat{\mathcal{H}}_{int} = -t\sum_{<ij>,i\in A}(a_i^\dagger f_j^\dagger h_i + h.c.) + V^*d\sum_{i\in B} f_i^\dagger f_i$.

The entire linearized Hamiltonian (1), using the the Bogoliubov transformation, takes the following form:

$$\mathcal{H}_L = \sum_{\mathbf{k}}[\epsilon_1 h^\dagger_{\mathbf{k}} h_{\mathbf{k}} + \epsilon_2 f^\dagger_{\mathbf{k}} f_{\mathbf{k}}] + \sum_{\mathbf{k},\mathbf{q}}\Big[g^{(1)}_{\mathbf{kq}}(f^\dagger_{\mathbf{k}-\mathbf{q}} h_{\mathbf{k}} \alpha^\dagger_{\mathbf{q}} + h^\dagger_{\mathbf{k}} f_{\mathbf{k}-\mathbf{q}} \alpha_{\mathbf{q}}) + g^{(1)}_{\mathbf{kq}}(f^\dagger_{\mathbf{k}} h_{\mathbf{k}-\mathbf{q}} \beta_{\mathbf{q}} + h^\dagger_{\mathbf{k}-\mathbf{q}} f_{\mathbf{k}} \beta^\dagger_{\mathbf{q}})\Big] + \mathcal{H}^B_L,$$

where $\epsilon_1 = 0$, $\epsilon_2 = V^* d$ and H^B_L is given by Eq. (6). The operators $f^\dagger_{\mathbf{k}}$ and $h^\dagger_{\mathbf{k}}$ are the Fourier transforms of $f^\dagger_i$ and $h^\dagger_i$ respectively and where $g^{(1)}_{\mathbf{kq}} = d\sqrt{2/N} t \gamma_{\mathbf{k}-\mathbf{q}} u_{\mathbf{q}}$, $g^{(2)}_{\mathbf{kq}} = d\sqrt{2/N} t \gamma_{\mathbf{k}} v_{-\mathbf{q}}$. The two Dyson's equations in the non-crossing approximation are given as follows [14]:

$$G_\nu(\mathbf{k},\omega) = \frac{1}{\omega - \epsilon_\nu - \sum_{\mathbf{q}} g^{(\nu)2}_{\mathbf{kq}} G_{\nu'}(\mathbf{k}-\mathbf{q}-\omega-\omega^\mu_{\mathbf{q}})}, \quad (7)$$

where the first equation is obtained for $\nu = 1$, $\nu' = 2$ and $\mu = \alpha$ and the second equation for $\nu = 2$, $\nu' = 1$ and $\mu = \beta$. Here $\omega_{\alpha,\beta}(\mathbf{q})$ are given after Eq. 6. The Green's function G_1 (G_2) corresponds to the quasi-particles created by $h^\dagger_{\mathbf{k}}$ ($f^\dagger_{\mathbf{k}}$). These equations can be solved iteratively as in Ref. [14] starting from $G^{(0)}_\nu(\mathbf{k},\omega) = 1/(\omega - \epsilon_\nu + i\eta)$.

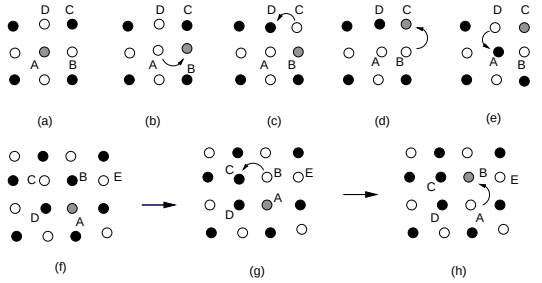

Fig. 2: The substitutional impurity moves by means of the 4^{th} order process (a)-(e). The interstitial impurity moves via the 2^{nd} order process (f)-(h). The impurity, bosonic atoms, and empty interstitial sites are denoted as gray solid, black solid, and open circles respectively.

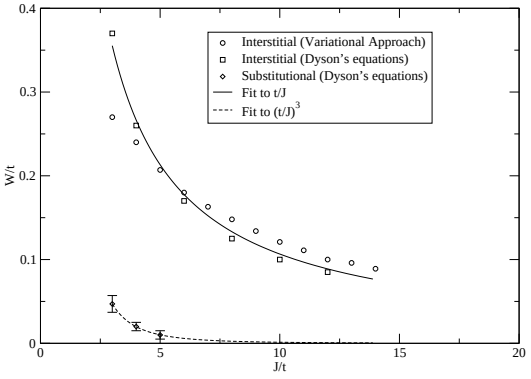

Fig. 3: The bandwidth of the substitutional and interstitial impurity bands as a function of J/t.

In our calculations we took $V^* = V = J$, $J_{xy} = 2t$ (i.e., $t = t_b$), $H = 0$ and $\epsilon = 0.1$. As discussed later NMR measurements [15] indicate that for ^3He impurities in solid ^4He we should consider $J/t >> 1$. In Fig. 1 the spectral function $A(\mathbf{k} = 0, \omega)$ is presented for the case of $J/t = 3$ (left) and $J/t = 6$ (right). Notice that for $J/t = 3$ the bandwidth W_A of the substitutional impurity is small. For $J/t = 6$, W_A is very small, i.e., $W_A/t \sim 10^{-3}$, and, hence, we only show the spectral function of the interstitial impurity for this value of J/t (right part of Fig. 1). Notice that the spectral function of the interstitial impurity has two main peaks, a lower frequency peak with small spectral weight and a higher frequency one with most of the spectral weight.

In the regime of $J >> t$, the leading order in t/J which allows the substitutional impurity or a ^4He atom to move is the fourth order process shown in Fig. 2(a-e). Hence, the impurity moves in a band with a bandwidth of the order of $W_A/t = A(t/J)^3$ in an expansion of t/J and J_{xy}/J (which is also small since $J_{xy} = 2t$). On the other hand, in our case where $V^* = V = J$, the bandwidth of the interstitial impurity is of the order of $W_B/t \sim t/J$ and it corresponds to the process shown in Fig. 2(f-h). States such as those of Fig. 2(f) and Fig. 2(h) are connected by second order degenerate perturbation theory processes where the states shown in Fig. 2(g) are included as intermediate states. Namely, the interstitial impurity at site A takes advantage of a pair "pseudo-spin" flip near it, as in Fig. 2(g), and hops to site B as shown in Fig. 2(h).

We also carried out a diagonalization in a space which includes the 17 states of the type shown in Fig. 2(f-h) and their translations through the square lattice. We found the dispersion indicated with the black and green lines in Fig. 1. The bandwidth of the impurity as calculated both by solving the Dyson's Eqs. 7 and by the variational approach is shown in Fig. 3 as a function of J/t. Notice that the interstitial band (Fig. 1) and bandwidth agree very well with those obtained from the Dyson's equations. The solid line and dashed line are fits to $W_B/t = B(t/J)$ and $W_A/t = A(t/J)^3$ respectively with $A \simeq B \simeq 1$.

Now, let us turn our discussion to the real case of ^3He impurities in solid ^4He. The form for bandwidth of the substitutional impurity, i.e., $W_A \sim t(t/J)^3$ (when we take $t \sim t_b$ and $V^* \sim V$), is also valid on the triangular and hcp lattices; namely, when the substitutional band is filled, in order for the atoms to move, the same fourth order process, where the atoms momentarily hop to interstitial positions, is necessary. In NMR [15] studies tunneling rates were found to be of the order of 1 MHz. Using our calculated form for the bandwidth $W_A = At(t/J)^3$, and taking $t = 1K$ and $J = 30K$, we find $W_A \sim 4 \times 10^{-5} K$ (i.e., 1 MHz). Using our form for the bandwidth for interstitial impurities $W_B = Bt^2/J$ and the same value of t and J we find $W_B \sim 40mK$. Hence, the present theory can reproduce (a) the NMR [15] results, (b) that substitutional quasiparticles might be localized [11] is solid ^4He, and (c) it suggests that *interstitial* impuritons might move coher-

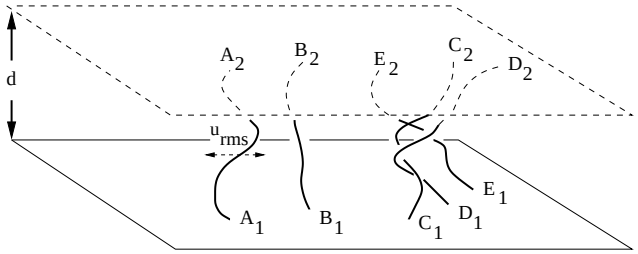

Fig. 4: Edge-dislocations or disclication lines form as the ^4He solid grows on the cell surface (bottom plane). The top (dashed) plane indicates the solid/liquid interface. We imagine that the solid grows layer by layer. Because the interface is two-dimensional, dislocations and disclinations might be expected to appear during a relatively fast cooling process at any given layer. In addition, the impurities present in the liquid bind very quickly to such point defects. The point defects of the next layer more or less line up with those of the previous layer (otherwise it is energetically very costly). Therefore, these defects become lines of defects as shown in the above figure. When the average distance between such defects is smaller than the length u_{rms} which characterizes the wandering of each defect line, the defect-lines can entangle (such as the lines C_1C_2, D_1D_2 and E_1E_2) and can produce a topological glass phase.

ently near the above temperature scale. However, as we discuss below, interstitial impurities bind with defects.

For the case of interstitial ^4He atoms, using our result for the bandwidth, $W = dJ_{xy}^2/2J$ (and $J_{xy} \sim 2t$), and the values of the parameters discussed in the previous paragraph, we find $W \sim 200mK$. This is the temperature scale where the possible super-solidity of ^4He has been observed [1]. In the case of real solid ^4He nearest neighbor empty interstitial sites are much closer to one another than occupied sites and, in addition, an interstitial atom has higher energy than a substitutional atom and therefore faces a lower potential barrier to tunnel to another interstitial site. These two factors increase the tunneling frequency by several orders of magnitude relative to the observed frequency [15] for substitutional sites. However, a finite energy is needed in order to promote atoms to the interstitial band [16] and, in addition, vacancies and interstitials have a tendency to phase separate in an equilibrated crystal [17]. On the other hand, inhomogeneities provided by ^3He impurities during the ^4He solid nucleation process or by a relatively fast inhomogeneous cooling process can create such interstitial atoms as discussed next.

As already discussed, the model studied in this paper is very similar to the $t-J$ model used to understand quantum antiferromagnets, where the impurity degrees of freedom in our case map to the holes in the $t-J$ model and the hopping of the bosonic atoms maps to the spin degrees of freedom. In the case of the $t-J$ model on the square lattice, when a finite density of holes is introduced the ground state may have stripe-order [18] where the stripes are hole-rich and the domains between stripes are antiferromagnetically ordered (quantum solid domains) with a π-phase shift between domains. As we will argue below, a similar phenomenon should be expected to occur in the case of an hcp lattice of bosons with impurities. Namely, edge-dislocations or disclination lines such as those shown in Fig. 4 might be created during the process of solid ^4He growth from the liquid phase.

First, it is known that the inhomogeneous nucleation of solid ^4He starts on the cell walls [19]. We will assume that solid ^4He grows layer-by-layer on the surface of the cell for the following reasons: (a) The geometry of the cells used in the experiments [1, 20] is characterized by a large surface with small distances between large confining walls. (b) The cooling of the solid is caused through cooling of these large surfaces, therefore, atoms near the surface will get cold first. (c) Helium atoms at temperature $T \sim 1K$ get adsorbed on the surface of most substrates because of a relatively strong dipolar interaction with the surface [21]. In studies of ^4He on graphite [22,23] for example and in all known substrates the first layer of adsorbed ^4He is solid. The second layer of ^4He on the graphite surface is also solid at layer completion density [22, 24]. After the first or a few layers are deposited on the surface, the adsorbed solid ^4He layer is *compressed* relative to the liquid in contact and, therefore, the coated surface provides a stronger attractive potential for an atom as compared to that provided by its surrounding atoms in the liquid; thus, the atoms tend to get adsorbed and form a new solid layer. Nucleation of solid ^4He had been a puzzle in the past just because of the large surface energy cost required in homogeneous nucleation [19]. In a layer-by-layer growth this is not an issue, because when the next layer is deposited, there is no additional surface area introduced, namely, just the same-size interface advances.

It is believed [25] that the solid/liquid interface in the presence of ^3He impurities becomes rough due to binding of such impurities to the interface. Even an extremely low impurity concentration, namely as low as 1 ppm, has a significant effect in this process [25]. In this paper, it is proposed that the ^3He impurities bind to defects [26], namely to dislocations or disclinations, which are expected to form on the two-dimensional solid/liquid interface [27]. There are various theories of two dimensional melting [28], the most popular of which is the theory of Halperin, Nelson and Young [27] which was inspired by the Kosterlitz-Thouless(KT) theory of vortices in superfluid films. According to this theory, the melting proceeds via two continuous KT transitions, the lowest temperature one caused by the unbinding of dislocations and a higher temperature transition caused by the unbinding of disclinations. Both types of defects are point-like singularities; the former corresponds to the singular behavior of the atomic lattice displacement field and the latter is associated with the singular behavior of the angle field $\theta(\vec{r})$ which characterizes the fluctuations in the bond orientational order. Our following analysis applies equally to both type of de-

fects and, therefore, we will use the term *defect* in general. As the solid/liquid interface is cooled down at a relatively fast rate, locally small-size 2D clusters of atoms at the solid/liquid interface achieve local crystalline order rather quickly; however, the time scale to achieve a global equilibrium is much longer because these clusters may have different orientations. In order to describe the state of this globally disordered but locally ordered 2D system, we need to imagine the presence of dislocations (or disclinations). Generally, as a 2D disordered solid is cooled down to low temperature, pairs of such thermally created excitations having opposite topological charge move very slowly towards each other and are annihilated (or bind). However, the ^3He impurities which are present in the liquid find it energetically favorable to bind to these defects and the combined impurity/defect system becomes stable. Furthermore, it becomes energetically favorable for these lattice defects of the next deposited solid layer to follow the positions of the defects on the previous layer, thus, defect-lines are formed as the interface advances; namely, lines of singularities grow perpendicular to the interface (see Fig. 4).

The root-mean-square projection u_{rms} of the end-to-end vector of a defect-line is given as [29] $u_{rms} = \sqrt{< u^2 >} = (2\pi k_B T d/\epsilon)^{1/2}$ where d is the thickness of the solid film and ϵ is the line tension along the defect-line. When u_{rms} is a few times greater than the average distance between such defects, neighboring defect-lines will entangle. Taking $T \sim 1K$ and $\epsilon \sim 10K/\mathring{A}$ and $d = 0.3mm$ we find that $u_{rms} \sim 10^3 \mathring{A}$ which is greater than the average distance between impurities and, thus, entanglement seems likely. When the solid is cooled to low temperature defects with opposite topological charge cannot be mutually annihilated because the entangled defects cannot move. The entanglement is topologically protected and this could create a topological glass state of solid ^4He. This is analogous to the vortex-glass state proposed for superconductors [30, 31].

As discussed earlier ^3He impurities, even at concentrations as low as 1 ppm, influence very significantly the solid ^4He nucleation process [25]. In particular even such very small amount of impurities can change the roughening transition temperature [25] by 20%. This has been interpreted as the result of ^3He impurity adsorption at the liquid to solid interface. As discussed such impurities stabilize the defects and as the interface advances defect lines form which may entangle and this can influence significantly the roughening transition. Therefore, it is possible that the ^3He impurities, through the creation of such defects, promote ^4He atoms to the interstitial band which can move from domain to domain and by means of the entangled defects. This creates a metastable disordered supersolid in which the carriers flow through a topological glass of entangled defect-lines. This scenario might explain the observed hysteresis [2] by annealing of the solid helium sample. Most recently Rittner and Reppy [20] have observed very high superfluid response in their torsional oscillator experiments of solid ^4He samples grown from the liquid phase by very rapid cooling. Furthermore, these samples have high surface-to-volume-ratio and high concentration of such defects is expected to occur under such conditions. Namely, under rapid cooling such defects are expected to occur in the 2D solid/liquid interface, because, the atoms order locally to form a microscopic-size solid very quickly, but the time scale for the annihilation of pair of defects of opposite topological charge is very long, namely much longer than the time required to establish local (i.e., at a microscopic scale) equilibrium. Once these defect-lines are formed, they wander around as shown in Fig. 4 and they may entangle around each other several times because of their high density. The process of annihilation of these defects is then very difficult because of their entanglement and this may lead to an extremely long-lived topological glass phase.

Experimentally, while the critical temperature T_c increases with increasing ^3He impurity concentration x_3, for the superfluid response ρ_s there is an optimum x_3 above which ρ_s decreases with increasing x_3. This behavior is consistent with the theory presented here. The degree of inhomogeneity is proportional to the density of defects which is also proportional to the impurity concentration x_3. The carrier density, namely the density of interstitial atoms, increases with x_3, therefore, T_c is expected to increase with x_3. Since T_c is more or less a locally determined quantity, the effect of global disorder on T_c is much less important than the fact that the number of carriers rise with disorder. The superfluid response, however, is expected to depend strongly on phase fluctuations of the superfluid order parameter, therefore, disorder is expected to have harmful effects to the long-range coherence. In summary, it is reasonable to expect that at high concentration of such ^3He impurities, the disorder which is caused by the lattice defects should significantly harm the long-range coherence, however, T_c may continue to rise because, while the long-range disorder does not significantly influence the value of T_c, T_c increases due to the increase of the number of carriers.

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Possible role of ^3He impurities in solid ^4He

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We use a quantum lattice gas model to describe the essential aspects of the motion ^4He atoms and of a ^3He impurity in solid ^4He. We find that ^3He impurities promote ^4He atoms to interstitial sites and this can turn the bosonic quantum crystal into a metastable supersolid. It is suggested that ^3He impurity atoms, which produce the interstitial ^4He atoms, might have been reabsorbed by pure ^4He solid formed during the first stage of a multi-stage solid ^4He nucleation process. While we use the "spin"-wave approximation and low dimensional lattices to illustrate some of the ideas, we argue that the conclusions drawn from these studies may be valid for the real system.

Kim and Chan[1, 2] (KC) using the torsional oscillator technique found a decrease in the resonant period of solid ^4He confined in porous vycor glass[1] and in bulk solid helium[2] below $200mK$, indicating the possible onset of superfluidity in solid helium. The experimental results of KC have been independently confirmed by Rittner and Reppy[3] (RR) using a different geometry to confine the solid. In addition, RR observed that the superfluidity of solid helium can be significantly influenced or eliminated by annealing of the solid helium sample. Because of these history-dependent results and the negative results in attempts to drive flow by pressure[4], the interpretation of the results of KC is subject to debate.

Furthermore, it has been observed that the superfluid response and superfluid fraction are strongly dependent on the amount of isotopic ^3He impurities which exist in the naturally available helium. When, for example, the naturally occurring concentration of ^3He impurities of about one part per million is reduced to less than one part per billion[5] the superfluid fraction is reduced from about 1% to approximately 0.03%. When the ^3He impurity concentration is increased to about 0.1% the superfluid fraction vanishes[1]. Previously, Ho *et al.*[6] found an anomalous behavior of the acoustic attenuation and velocity in solid ^4He below $\sim 200mK$ at a low concentration of ^3He impurities.

The possibility of superfluidity of solid ^4He has been extensively discussed[7, 8, 9, 10, 11]. It is of fundamental value for our understanding of this and related phenomena because of the implication that there can be coexistence between spatial and momentum space order[12].

In this paper we study the role of very low density of impurities in a bosonic hard core solid. While the theoretical studies of the role of impurities in solid helium has a long history[7, 13, 14], the present study, based on an analogy with models of doped quantum antiferromagnets and by using rather recently developed techniques for such systems, sheds some "new" light on the problem. Our model, that describes the impurity motion in an otherwise ideal quantum bosonic crystal, maps to a quantum spin model with antiferromagnetic (AF) coupling and AF order in one direction and ferromagnetic coupling in the perpendicular direction with impurities moving through the lattice. The impurity motion between sub-lattices couples to quantum fluctuations of these pseudo-spin degrees of freedom which correspond to the boson hopping from an occupied site of the solid to an empty interstitial site. We find that substitutional impurities become localized and an interstitial impurity relaxes into a substitutional site by promoting a ^4He atom to the interstitial band. Furthermore, substitutional ^3He impurities, because of their larger zero point motion, tend to promote ^4He atoms into the interstitial band. We discuss that the bandwidth of the interstitial band is of the same order of magnitude as the temperature scale below which the non-classical moment of inertia was observed[1, 2].

Let us consider a lattice gas model to describe the bosonic solid and the added impurities. In such a model, we need to consider the interstitial sites as part of the lattice and, thus, the ideal quantum solid containing no vacancies and no impurities, corresponds to a fractionally occupied lattice. For example, the ideal triangular solid corresponds to the case of 1/3 filling, namely, to a $\sqrt{3}\times\sqrt{3}$ ordered solid and the ideal square lattice solid corresponds to the $\sqrt{2}\times\sqrt{2}$ checkerboard solid, i.e., 1/2 filling of the lattice with bosons. Our model Hamiltonian describing a bosonic quantum solid (such as solid ^4He) and a small concentration of impurities (such as ^3He atoms) may be written as follows:

$$\hat{\mathcal{H}} = \hat{\mathcal{H}}_B + \hat{\mathcal{H}}^* + \hat{\mathcal{H}}_{int} - \mu\sum_i \hat{N}_i - \mu^*\sum_i \hat{n}_i, \quad (1)$$

$$\hat{\mathcal{H}}_B = \sum_{<ij>}[-t_b(B_i^\dagger B_j + h.c.) + V\hat{N}_i\hat{N}_j] + U\sum_i \hat{N}_i^2, \quad (2)$$

$$\hat{\mathcal{H}}^* = \sum_{<ij>}[-t(c_i^\dagger c_j + h.c.) + W\hat{n}_i\hat{n}_j] + U^*\sum_i \hat{n}_i^2, \quad (3)$$

$$\hat{\mathcal{H}}_{int} = V^*\sum_{<ij>}(\hat{N}_i\hat{n}_j + \hat{n}_i\hat{N}_j) + U'\sum_i \hat{n}_i\hat{N}_i, \quad (4)$$

$\mathcal{H}_B$ is the Hamiltonian of the bosonic solid, and $B_i^\dagger(B_i)$ are boson creation (annihilation) operators and $\hat{N}_i = B_i^\dagger B_i$. The fermion operators $c_i^\dagger(c_i)$ create (or annihilate) impurities on the site i and we have suppressed their spin

degree of freedom for simplicity. Here, n_i is the impurity number operator. We will consider the $U \to \infty$, $U^* \to \infty$ and $U' \to \infty$ limits (single-site occupation subspace) because of the the hard-core interaction. V, V^* and W are positive because of the additional energy cost to place an atom in an interstitial site.

In the absence of impurities the well-known pure bosonic Hamiltonian (2), in the limit of $U \to \infty$ and under the transformation $B_i^\dagger \to S_i^+, B_i \to S_i^-, N_i \to S_i^z - \frac{1}{2}$, where $(\hat{S}_i^x, S_i^y, S_i^z)$ are spin-1/2 operators, reduces to the anisotropic spin-1/2 Heisenberg model[15]

$$\hat{\mathcal{H}}_B = \sum_{<ij>} [J S_i^z S_j^z + J_{xy}(S_i^x S_j^x + S_i^y S_j^y)] - H \sum_i S_i^z, \quad (5)$$

where $J = V > 0$ and $J_{xy} = -2t_b < 0$ and $H = \mu - z/2V$, z is the lattice site coordination number.

In order to illustrate the effects of the impurities on the stability of the quantum solid and, vice versa, we first consider the square lattice because of its simplicity. Following the general spin-wave (SW) theory for an ordered square lattice quantum antiferromagnet[15], we separate the ordered square lattice in two sub-lattices, A (or up, or occupied for the case of the solid) and B (down, or empty) and we consider boson operators $a_i^\dagger$ and $b_i^\dagger$ which create spin-deviations with respect to the classical Néel ground state in sites of the corresponding sub-lattice. The Hamiltonian (5) is approximated by keeping terms up to quadratic in spin-deviation operators; using the Fourier transforms $a_{\mathbf{k}}$ and $b_{\mathbf{k}}$ of the operators a_i and b_i (as defined in Ref. 15), where $\mathbf{k}$ takes values from the Brillouin zone of the $\sqrt{2} \times \sqrt{2}$ sub-lattice and introducing the Bogoliubov canonical transformation, $a_{\mathbf{k}} = u_k \alpha_{\mathbf{k}} + v_k \beta_{-\mathbf{k}}^\dagger$, $b_{\mathbf{k}} = u_k \beta_{\mathbf{k}} - v_k \alpha_{-\mathbf{k}}^\dagger$, where $\alpha_{\mathbf{k}}^\dagger$ and $\beta_{\mathbf{k}}^\dagger$ are boson creation operators, the quadratic linearized Hamiltonian can be diagonalized and it takes the form

$$\mathcal{H}_L^B = E_0 + \sum_{\mathbf{k}} \Big(\omega_{\mathbf{k}}^\alpha \alpha_{\mathbf{k}}^\dagger a_{\mathbf{k}} + \omega_{\mathbf{k}}^\beta \beta_{\mathbf{k}}^\dagger \beta_{\mathbf{k}} \Big), \quad (6)$$

where $\omega_{\mathbf{k}}^{\alpha,\beta} = dJ\epsilon_{\mathbf{k}} \pm H$, $\epsilon_{\mathbf{k}} = \sqrt{1 - \lambda^2 \gamma_{\mathbf{k}}^2}$, $\gamma_{\mathbf{k}} = 1/2(cos(k_x) + cos(k_y))$, $d = 2$ and $\lambda = J_{xy}/J$, provided that we choose the functions $u_{\mathbf{k}}$ and $v_{\mathbf{k}}$ as follows: $u_{\mathbf{k}} = \sqrt{1/2(1/\epsilon_{\mathbf{k}} + 1)}$, $v_{\mathbf{k}} = -sgn(\gamma_{\mathbf{k}})\sqrt{1/2(1/\epsilon_{\mathbf{k}} - 1)}$.

It follows from Eq. 6 that for $\omega_0^{\alpha,\beta} \geq 0$, i.e., for $H \geq dJ\sqrt{1-\lambda^2}$, the Néel ordered ground state is unstable. Ferromagnetic (superfluid in the bose system) order develops in the xy direction and the spins are canted in order to acquire a component along the direction of the field. The phase diagram obtained for this model with this spin-wave approximation agrees reasonably well with that obtained by other techniques.[16] When the square lattice is half-filled there is a gap $G = dJ\sqrt{1-\lambda^2}$ for creating a propagating pseudo-spin wave excitation, i.e., to promote a boson atom to the interstitial band. These interstitial quasiparticles move in a band which in the limit of $J_{xy} << J$ has a bandwidth $W = dJ_{xy}^2/2J$.

We wish to extend this approach to study the motion of a single impurity inside the quantum solid. As in Ref. 17, we consider as reference state the Néel state with an impurity on the A (substitutional) or B (interstitial) sublattices. We imagine that there exist operators $h_i^\dagger$ and $f_i^\dagger$ which operate on the Néel state and replace respectively an up-spin or down-spin with an impurity. For the single impurity case and by keeping only up to linear terms in spin-deviation operators the terms given by Eqs. (3) (4) in terms of these operators take the form $\hat{\mathcal{H}}^* + \hat{\mathcal{H}}_{int} = -t \sum_{<ij>, i \in A} (a_i^\dagger f_j^\dagger h_i + h.c.) + V^* d \sum_{i \in B} f_i^\dagger f_i$.

The entire Hamiltonian (1) when linearized and for the case of a single impurity, as described above, and by using the Bogoliubov transformation for the boson operators, takes the following form:

$$\mathcal{H}_L = \sum_{\mathbf{k}} [\epsilon_1 h_{\mathbf{k}}^\dagger h_{\mathbf{k}} + \epsilon_2 f_{\mathbf{k}}^\dagger f_{\mathbf{k}}] + \sum_{\mathbf{k},\mathbf{q}} \Big[g_{\mathbf{kq}}^{(1)} (f_{\mathbf{k}-\mathbf{q}}^\dagger h_{\mathbf{k}} \alpha_{\mathbf{q}}^\dagger + h_{\mathbf{k}}^\dagger f_{\mathbf{k}-\mathbf{q}} \alpha_{\mathbf{q}}) + g_{\mathbf{kq}}^{(1)} (f_{\mathbf{k}}^\dagger h_{\mathbf{k}-\mathbf{q}} \beta_{\mathbf{q}} + h_{\mathbf{k}-\mathbf{q}}^\dagger f_{\mathbf{k}} \beta_{\mathbf{q}}^\dagger) \Big] + \mathcal{H}_L^B,$$

where $\epsilon_1 = 0$, $\epsilon_2 = V^* d$ and H_L^B is given by Eq. (6). The operators $f_{\mathbf{k}}^\dagger$ and $h_{\mathbf{k}}^\dagger$ are the Fourier transforms of $f_i^\dagger$ and $h_i^\dagger$ respectively and where $g_{\mathbf{kq}}^{(1)} = d\sqrt{2/N} t \gamma_{\mathbf{k}-\mathbf{q}} u_{\mathbf{q}}$, $g_{\mathbf{kq}}^{(2)} = d\sqrt{2/N} t \gamma_{\mathbf{k}} v_{-\mathbf{q}}$. The two Dyson's equations in

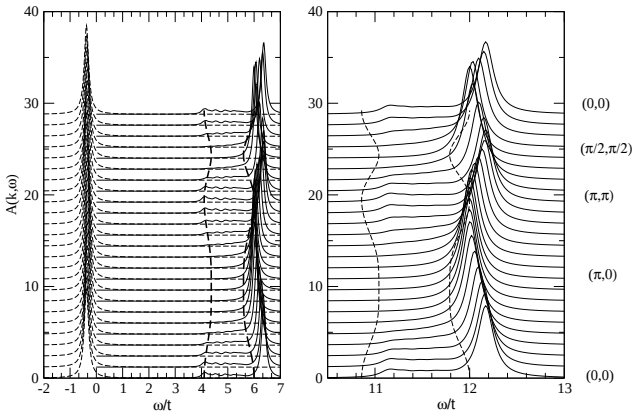

FIG. 1: The spectral functions $A(\mathbf{k}, \omega)$ for $J/t = 3$ (left) and $J/t = 6$ (right) along the Brillouin zone path $(0.0) \to (\pi, 0) \to (\pi, \pi) \to (0, 0)$.

the non-crossing approximation are given as follows[17]:

$$G_\nu(\mathbf{k}, \omega) = \frac{1}{\omega - \epsilon_\nu - \sum_{\mathbf{q}} g_{\mathbf{kq}}^{(\nu)2} G_{\nu'}(\mathbf{k} - \mathbf{q} - \omega - \omega_{\mathbf{q}}^\mu)}, \quad (7)$$

where the first equation is obtained for $\nu = 1$, $\nu' = 2$ and $\mu = \alpha$ and the second equation for $\nu = 2$, $\nu' = 1$ and $\mu = \beta$. Here $\omega_{\alpha,\beta}(\mathbf{q})$ are given after Eq. 6. The Green's function G_1 (G_2) corresponds to the quasi-particles created

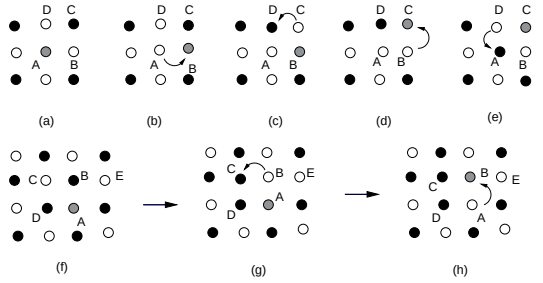

FIG. 2: The substitutional impurity moves by means of the 4^{th} order process (a)-(e). The interstitial impurity moves via the 2^{nd} order process (f)-(h). The impurity, bosonic atoms, and empty interstitial sites are denoted as gray solid, black solid, and open circles respectively.

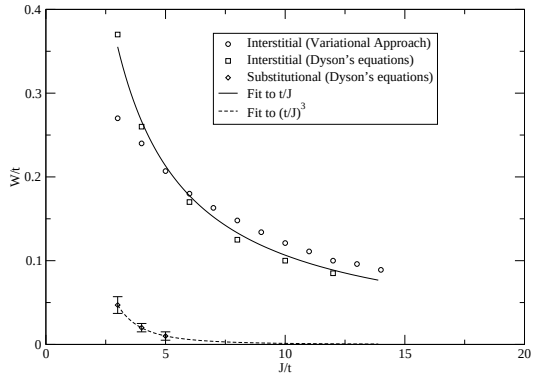

FIG. 3: The bandwidth of the substitutional and interstitial impurity bands as a function of J/t.

by $h^\dagger_{\mathbf{k}}$ ($f^\dagger_{\mathbf{k}}$). These equations can be solved iteratively as in Ref. 17 starting from $G^{(0)}_\nu(\mathbf{k},\omega) = 1/(\omega - \epsilon_\nu + i\eta)$.

In our calculations we took $V^* = V = J$, $J_{xy} = 2t$ (i.e., $t = t_b$), $H = 0$ and $\epsilon = 0.1$. As discussed later NMR measurements[18] indicate that for ^3He impurities in solid ^4He we should consider $J/t >> 1$. In Fig. 1 the spectral function $A(\mathbf{k} = 0, \omega)$ is presented for the case of $J/t = 3$ (left) and $J/t = 6$ (right). Notice that for $J/t = 3$ the bandwidth W_A of the substitutional impurity is small. For $J/t = 6$, W_A is very small, i.e., $W_A/t \sim 10^{-3}$, and, hence, we only show the spectral function of the interstitial impurity for this value of J/t (right part of Fig. 1). Notice that the spectral function of the interstitial impurity has two main peaks, a lower frequency peak with small spectral weight and a higher frequency one with most of the spectral weight.

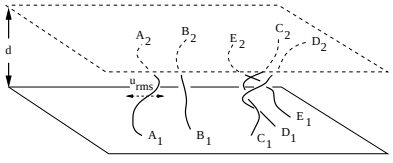

FIG. 4: A substitutional ^3He impurity in solid ^4He promotes ^4He atoms from the substitutional to the interstitial band. The size of the black circles is proportional to the ^4He occupancy probability. The white circles indicate empty interstitial sites. The site A is occupied by a ^3He impurity.

In the regime of $J >> t$, the leading order in t/J which allows the substitutional impurity or a ^4He atom to move is the fourth order process shown in Fig. 2(a-e). Hence, the impurity moves in a band with a bandwidth of the order of $W_A/t = A(t/J)^3$ in an expansion of t/J and J_{xy}/J (which is also small since $J_{xy} = 2t$). On the other hand, in our case where $V^* = V = J$, the bandwidth of the interstitial impurity is of the order of $W_B/t \sim t/J$ and it corresponds to the process shown in Fig. 2(f-h). The states such as those of Fig. 2(f) and those of Fig. 2(h) are degenerate and are connected by second order degenerate perturbation theory processes where the states shown in Fig. 2(g) are included as intermediate states. Namely, the interstitial impurity at site A takes advantage of a pair "pseudo-spin" flip near it, as in Fig. 2(g), and hops to site B as shown in Fig. 2(h).

We also carried out a diagonalization in a space which includes the 17 states of the type shown in Fig. 2(f-h) and their translations through the square lattice. We found the dispersion indicated with the black and green lines in Fig. 1. The bandwidth of the impurity as calculated both by solving the Dyson's Eqs. 7 and by the variational approach is shown in Fig. 3 as a function of J/t. Notice that the interstitial band (Fig. 1) and bandwidth agree very well with those obtained from the Dyson's equations. The solid line and dashed line are fits to $W_B/t = B(t/J)$ and $W_A/t = A(t/J)^3$ respectively with $A \simeq B \simeq 1$.

Now, let us turn our discussion to the real case of ^3He impurities in solid ^4He. The form for bandwidth of the substitutional impurity, i.e., $W_A \sim t(t/J)^3$ (when we take $t \sim t_b$ and $V^* \sim V$), is also valid on the triangular and hcp lattices; namely, when the substitutional band is filled, in order for the atoms to move, the same fourth order process, where the atoms momentarily hop to interstitial positions, is necessary. In NMR[18] studies tunneling rates were found to be of the order of 1 MHz. Using our calculated form for the bandwidth $W_A = At(t/J)^3$, and taking $t = 1K$ and $J = 30K$, we find $W_A \sim 4 \times 10^{-5} K$ (i.e., 1 MHz). Using our form for the bandwidth for interstitial impurities $W_B = Bt^2/J$ and the same value of t and J we find $W_B \sim 40mK$. Hence, the present theory can reproduce (a) the NMR[18] results, (b) that substitutional quasiparticles might be localized[14] is solid ^4He, and (c) it suggests that *interstitial* impuritons might move coherently near the above temperature scale. However, as we discuss below interstitial impurities lighter than the ^4He atoms very quickly relax into substitutional sites.

For the case of interstitial ^4He atoms, using our result for the bandwidth, $W = dJ^2_{xy}/2J$ (and $J_{xy} \sim 2t$), and the

values of the parameters discussed in the previous paragraph, we find $W \sim 200mK$. This is the temperature scale where the possible super-solidity of ^4He has been observed[1, 2]. In the case of real solid ^4He nearest neighbor empty interstitial sites are much closer to one another than occupied sites and in addition, an interstitial atom has higher energy than a substitutional atom and therefore faces a lower potential barrier to tunnel to another interstitial site. These two factors increase the tunneling frequency by several orders of magnitude relative to the observed frequency[18] for substitutional sites. However, an energy gap is needed in order to promote them to the interstitial band and the role of ^3He impurities becomes important right here as discussed next.

An interstitial impurity is expected to quickly relax to a substitutional site by promoting a ^4He atom to an interstitial position. The reason is that the ^3He atomic mass is smaller than the ^4He mass, therefore, the configuration in which the ^3He atoms is "squeezed" in the interstitial space has significantly higher energy than the one where the ^4He is in the interstitial space. For a similar reason even a substitutional ^3He atom has a tendency to promote a ^4He atom to an interstitial space as illustrated in Fig. 4. A substitutional ^3He impurity in solid ^4He prefers a larger room for zero-point motion than the solid ^4He lattice spacing because of its lighter mass. As a result, it causes a local expansion which means that the atoms labeled as 1-6 in the Fig. 4 will spend some time as interstitial ^4He atoms. This implies that ^3He impurities are expected to promote ^4He atoms from the substitutional band which is full to an empty interstitial band with a relatively large bandwidth.

Interstitial impurities and interstitial ^4He atoms could be created by the non-equilibrium nucleation process that produces solid ^4He from the liquid phase. The kinetics of nucleation and of phase separation of solid $^3He-^4He$ mixtures is still under experimental investigation[19, 20, 21]. It is known that there is a great asymmetry between the kinetics of phase separation and homogenization in solid $^3He-^4He$ mixtures. The observation of anomalously fast mass transfer at inclusion-dissolution of ^3He in solid ^4He matrix was explained by ballistic transfer of ^3He quasiparticles[21]. ^4He requires a lower pressure than ^3He to solidify; thus, it is possible that, at a first stage, the nucleation of pure ^4He takes place and the ^3He impurity atoms are excluded when solid clusters form. At a later stage these ^3He atoms may be slowly reabsorbed through diffusion inside the solid ^4He crystal, which, as discussed, produce interstitial ^4He atoms which should remain as metastable defects in the crystal. This scenario might explain the observed hysteresis[3] by annealing of the solid helium sample.

If indeed ^3He impurities drive the superfluid behavior in solid ^4He, the condensate fraction, naively, should be proportional to the ^3He impurity concentration. Therefore, the large difference between condensate fraction and the observed superfluid fraction needs to be explained. A large difference between these cannot be excluded because we have examples, such as liquid ^4He near solidification and at low temperature, where the condensate fraction is small and the superfluid fraction is almost 100%. However, a simple explanation from the present work cannot be provided and further experimental and theoretical work is needed. Some of the proposed ideas can be tested using path integral Monte Carlo simulation[22].

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